Center for Electrochemical Energy Science (CEES)
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Lead Institution: Argonne National Laboratory
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Mission*: to develop a fundamental understanding and robust control of the reactivity of electrified oxide interfaces, films and materials relevant to lithium-ion batteries.*

Energy storage is a strategic technology that enables the large-scale use and distribution of electrical energy as well as mobile technologies such as electric vehicles and portable electronic devices. Lithium-ion batteries (LIBs) are attractive because they offer inherently high energy densities through the conversion of electrical and chemical energy through electrochemical reactions. However, LIBs place severe demands on the performance of the active materials. These reactions rely on the reversible lithium insertion in the battery electrodes and associated redox reactions. Challenges include the need to control deleterious side reactions such as electrolyte decomposition at the electrode-electrolyte interface (e.g., solid-electrolyte interphase "SEI" formation), dissolution of the active materials, and the need to restructure the electrode framework materials at high lithium capacities.

The *Center for Electrochemical Energy Science* (CEES) explores the electrochemical reactivity of oxide materials that represent the positive electrode (e.g., cathode) in many lithium ion battery (LIB) systems. To this end, CEES seeks to *observe* and *understand* the reactivity of electrified oxide interfaces, films and materials, and to *control* these complex lithiation processes through the use of novel structures, architectures and chemistries. Such an understanding will define new conceptual approaches to prevent side-reactions, minimize over-potentials, and enable substantial improvements in energy density of the battery. In this way, the research program is designed to explore the fundamental limits of LIB technologies and to enable disruptive advances in energy storage systems.

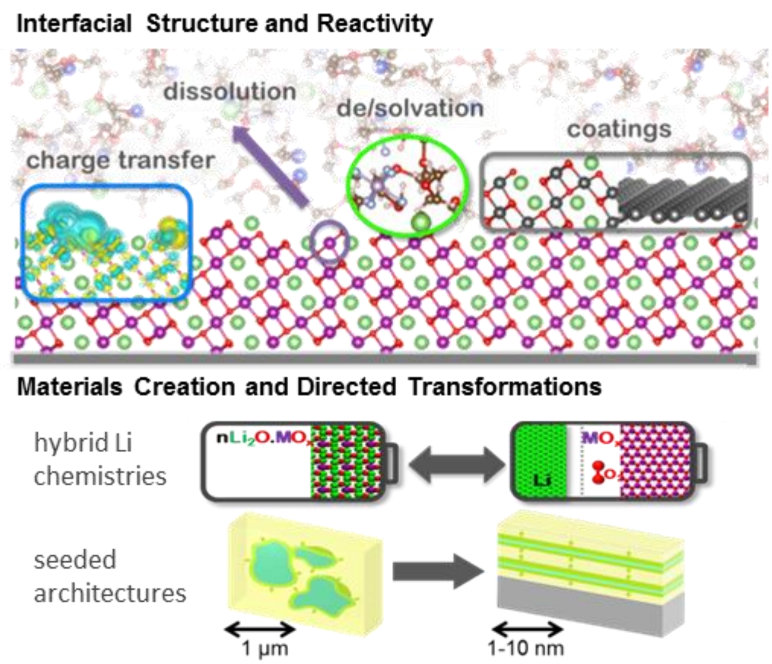

Figure: (Top) Schematic of interfacial structures and coatings to control secondary reactions at insertion electrodes interfaces. (Bottom) Directing chemical reactivity through the use of advanced architectures for conversion reactions and novel chemistries associated with hybrid Li^+/Li-O reactions.

To this end, CEES addresses two broad issues: (indicated schematically in the **Figure**).

- **Interfacial Structure and Reactivity**: *Can we control reactions at the oxide-electrolyte interface by leveraging a robust molecular-scale understanding of its structure and reactivity?* The transport of ions across the electrode/electrolyte interface leads to over-potentials, and side reactions (e.g., cathode dissolution at elevated potentials) associated with interface-specific reactions. These interfacial aspects of electrochemistry introduce substantial challenges, but simultaneously offer many avenues for controlling and guiding reactivity through interfacial modification.

- **Directed Transformations:** *Can we discover new approaches that can be used to direct the character and nature of electrochemical reactions within electrode materials?* Here, we seek to influence the pathway of an electrochemically-driven reaction within an electrode. There are two focus areas of research. In one subtask, we seek to use dimensionality to control conversion chemistries in the lithation of nano-structured electrodes. In a second subtask, we seek to explore the use of dual functioning electrode/electrocatalyst materials to enable reversible lithium and oxygen extraction from a lithium-transition metal-oxide crystal structure while allowing partial redox of the transition-metal and oxygen ions.

These guiding questions will be addressed using the unique and proven multi-pronged approach that CEES has established and used since 2009. To this end, CEES will probe the intrinsic reactivity of well-defined model oxide systems, with molecular-scale sensitivity and resolution, to reveal the inherent complexity of electrochemical systems (including the relevant active materials, electrolyte, etc.). The use of precisely defined materials, structures and interfaces (for example, structurally and compositionally defined epitaxial thin films) will enable direct observations of the relevant reactions without the complications of inactive materials and complex electrode morphologies inherent to conventional systems. This understanding, leveraged with the development of novel materials, architectures and chemistries, will be used to observe and direct electrochemical reactivity in these systems. These approaches, enabled by our deep cross-cutting capabilities of materials synthesis, characterization, and theory will lead to new approaches to control electrochemical reactions under the extreme conditions found in lithium-ion battery systems.

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